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Municipal solid waste incineration bottom ash supported cobalt oxide catalysts for dye degradation using sulfate radical

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ABSTRACT

Waste material of incineration bottom ash (IBA) was applied as support to load cobalt oxide catalysts for dye degradation using sulfate radicals. By using impregnation–calcination method, IBA supported cobalt oxide catalysts (Co/IBA) were prepared. Energy dispersive spectroscopy (EDS) showed that cobalt oxide particles were well dispersed on the surface of IBA. Catalytic activity evaluation showed that samples prepared using IBA with particle size of 0.212–1.4 mm exhibited similar high methylene blue (MB) degradation efficiencies, owing to the similar high Fe content in IBA particles. Catalytic reaction conditions were optimized in terms of MB degradation time. Besides MB, other dyes including rhodamine B (RhB), orange II sodium salt (OS) and malachite green (MG) could all be degraded within 10 min. In addition, recycle studies showed that the activity of Co/IBA catalyst was reasonably stable within three runs. These results demonstrate the potential application of IBA as low-cost and stable support material in heterogeneous catalysis.

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1. Introduction

Waste materials produced from both industrial and agricultural activities posed severe threat to environmental sustainability. The usage of waste materials as feedstocks has received increasing attention due to their abundance and low cost [1–6]. Apart from being used as landfilling materials, conversion of wastes to high-value-added products is of great importance for recycling and reusing of materials. Incineration bottom ash (IBA) generated from municipal solid waste treatment is one of the mostly investigated waste materials due to its huge amount and potential environmental risk [7–9]. The lack of landfill sites and certain toxic heavy metals (Pb, Cd, Cr, Cu, Zn, etc.) leaching into the surroundings pose threats to the environment. Although landfill is still the primary option for IBA disposal currently, many research efforts have been devoted to the conversion of IBA to high-value-added and environmentally benign products, such as cement [10], concrete [11], adsorbents [12] etc. The main components of IBA including silicon, calcium and aluminum oxide, are ideal for conversion to zeolitic-type and layered materials as adsorbents [13]. Zeolite, tobermorite, sodium aluminum silicate hydrate, katoite and sodium aluminum

phosphate silicate hydrate were produced from alkaline or neutral hydrothermal treatment of IBA. The IBA-converted products were applied as adsorbents for the removal of heavy metal ions and organic dye from water [12,14–18].

Dyes are important feedstocks in textile, paper and pulp, tannery and paint industries. It is estimated that about 1 million tons of dyes are produced annually, of which 10–15% used dyes enter the environment through water [19]. Due to the toxic nature of dyes to both human health and ecosystem, the release of colorant effluent to environment has become a major concern. Hence it is important to treat the colorant effluent to remove dyes before its release. Conventional strategies for dye removal from wastewater include adsorption [20], coagulation [21], filtration [22], ion exchange [23], microbiological treatment [24], electrochemical technique [25], photocatalysis [26], chemical oxidation [27] etc. In particular, sulfate radical-based advanced oxidation process (SR-AOP) is one of the most promising technologies for practical applications since it allows the complete destruction of dye molecules [28]. As a versatile and environmentally friendly oxidant, Oxone[®] with a molecular composition of $\text{KHSO}_5 \cdot 0.5\text{KHSO}_4 \cdot 0.5\text{K}_2\text{SO}_4$ is usually chosen to provide sulfate radicals after activation. Generally, Oxone[®] can be activated by energy system (thermal, photochemical or sonochemical) as well as catalyst system using transition metals [28]. Compared with energy systems which require high-cost equipment and high energy input, the catalyst system is more active and affordable hence it is highly preferred. Among various

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transition metal catalysts, the most active combination for sulfate radical generation was reported to be Co^{2+} coupling with Oxone[®] [29]. However, the potential health hazard of dissolved Co^{2+} in water limits the application of homogeneous catalyst system. Thus, heterogeneous catalyst systems using supported or unsupported cobalt oxide for Oxone[®] activation have been widely explored [30–36]. Particularly, it was found that the catalyst support played an important role in cobalt oxide/Oxone[®] activation system, which included dispersing cobalt oxide nanoparticles, minimizing the leaching of cobalt ions into liquid phase, and facilitating the formation of Co–OH complex [30]. Various support materials such as TiO_2 , Al_2O_3 , MgO , SiO_2 , graphene oxide, carbon nanosphere, metal–organic frameworks (MOFs), were used for cobalt oxide immobilization [30,33,35,36]. The supported cobalt oxide catalysts exhibited similar or even higher dye degradation efficiency than homogeneous Co^{2+} or unsupported cobalt oxide catalyst [30,33,34]. Typically, in a model aqueous solution using cobalt oxide/MgO as catalyst, methylene blue (MB) dye was degraded within 7 min, while 15 min was required using homogeneous Co^{2+} [30].

In this study, we aim to combine a model waste material of IBA with cobalt oxide to construct supported catalysts (denoted as Co/IBA). The Co/IBA was then used for dye degradation in aqueous solution using sulfate radicals. Compared with synthesized catalyst supports, our Co/IBA catalyst system has the advantage of low-cost due to the abundance of IBA. The loading of cobalt oxide on IBA was achieved by impregnation–calcination method. The produced Co/IBA catalysts were evaluated for dye degradation and demonstrated excellent activities. Effects of IBA particle size, cobalt loading amount, catalyst dosage, and Oxone[®] dosage on dye degradation efficiencies were systematically investigated. The optimized catalyst system of Co/IBA is expected to be a promising candidate for colorant effluent treatment.

2. Experimental section

2.1. Synthesis of IBA-supported cobalt oxide catalysts

Raw IBA samples collected from incineration plant were dried in an oven at 105 °C for 24 h upon sample collection. Sieving was conducted on a sieve shaker (Retsch, AS 200, Germany). Particles with a diameter of >1.4 mm, which mainly comprised of inorganic debris (metallics, ceramics, etc.), were discarded. According to mesh size, IBA particles (<1.4 mm) were separated to 4 particle size ranges of <0.212 mm (IBA-A), 0.212–0.6 mm (IBA-B), 0.6–1.0 mm (IBA-C), and 1.0–1.4 mm (IBA-D). All IBA samples were ball-milled for 2 h and then washed by de-ionized water before cobalt oxide loading.

IBA supported cobalt oxide catalysts were prepared by incipient wetness impregnation method [30]. Typically, 1 g of IBA-A was added to 20 ml aqueous solution containing 247 mg of $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$. The suspension was stirred at 80 °C until H_2O was totally evaporated. The obtained solid sample was dried at 60 °C overnight and then calcined at 600 °C for 3 h in static air. The adding amount of $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ was varied to study the effect of Co loading amount. The as-obtained sample was denoted as x%Co/IBA-A, where x indicates the amount of Co^{2+} added during the synthesis. The unsupported cobalt oxide catalyst was prepared by calcination of $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ at 600 °C for 3 h in static air, which was denoted as Co–C.

2.2. Characterization

Powder X-ray diffraction (XRD) patterns were obtained on a X-ray diffractometer (Bruker D2 Phaser, $\text{Cu K}\alpha$, $\lambda = 1.5406 \text{ \AA}$, 30 kV and 10 mA). The morphology of sample was observed on field emission scanning electron microscopy (FESEM, JEOL JSM6701F).

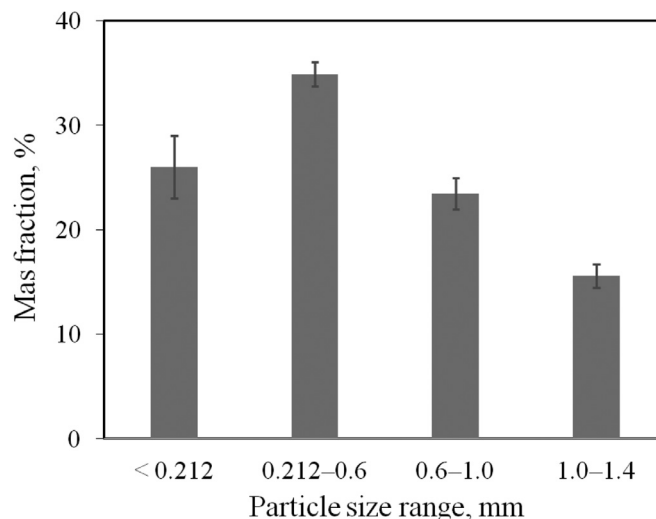


Fig. 1. Particle size distribution of IBA.

Energy dispersive spectroscopy (EDS, Oxford) equipped on FESEM was used to determine the elemental composition of sample. Nitrogen physisorption measurement was conducted at –196 °C on a Quantachrome Autosorb-6B apparatus. The specific surface area was determined using the Brunauer–Emmett–Teller (BET) method. UV-visible diffuse reflectance spectra (UV-vis DRS) were recorded on a UV-visible spectrophotometer (UV-2450, Shimadzu). The concentration of leached Co^{2+} ion concentration was measured by inductively coupled plasma optical emission spectroscopy (ICP-OES) on a Prodigy High Dispersion ICP (Leeman Teledyne) machine.

2.3. Catalytic activity evaluation

Dye degradation was conducted at room temperature in a glass beaker. To avoid the influence of indoor light, the beaker was covered with aluminum foil. In a typical run, 200 mg of catalyst and 0.4 mmol of Oxone[®] ($\text{KHSO}_5 \cdot 0.5\text{KHSO}_4 \cdot 0.5\text{K}_2\text{SO}_4$, Sigma) was added to 200 ml of methylene blue (MB) aqueous solution with constant stirring rate of at around 300 rpm. The starting concentration of MB was 0.1 mM. At certain reaction time intervals, 1 ml of suspension was withdrawn and mixed with 1 ml of NaNO_2 (5 g/l) immediately to quench the reaction. After centrifugation at 13 000 rpm to remove solid catalyst, the concentration of MB in the supernatant was measured on a UV-visible spectrophotometer (UV-2450, Shimadzu). The activity of Co/IBA catalyst was also evaluated for the degradation of rhodamine B (RhB), orange II sodium salt (OS) and malachite green (MG) under the same experimental conditions. For the recycle runs of MB degradation, the used catalyst was collected by centrifugation and washed thoroughly by de-ionized water and ethanol. After drying at 60 °C overnight, the catalyst was used for next run.

3. Results and discussion

3.1. Properties of IBA and Co/IBA catalysts

IBA was obtained from high temperature incinerator. Its major elemental compositions were determined to be SiO_2 , CaO , Fe_2O_3 and Al_2O_3 with the crystal phases of quartz, calcite, anorthite, gehlenite and potassium aluminum silicate hydrate as shown in our previous studies [13,18]. IBA with particle size larger than 1.4 mm, which contains mainly ceramic and metallic debris, was discarded (ca. 60 wt%). Fig. 1 shows the particle size distribution of leftover IBA (<1.4 mm, 40 wt%). It is found that 35 wt% of IBA

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